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Active Metamaterials with Negative Static Electric Susceptibility

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Although well-established textbook arguments suggest that static electric susceptibility $\chi^{(0)}$ must be positive in “all bodies”, it has been pointed out that materials that are not in thermodynamic equilibrium are not necessarily subject to this restriction. Media with inverted populations of atomic and molecular energy levels have been predicted theoretically to exhibit a $\chi^{(0)} < 0$ state, however the systems envisioned require reduced temperature, reduced pressure, and an external pump laser to maintain the population inversion. Further, the existence of $\chi^{(0)} < 0$ has never been confirmed experimentally. Here, a completely different approach is taken to the question of $\chi^{(0)} < 0$ and a design concept to achieve ‘true’ $\chi^{(0)} < 0$ is proposed based on active metamaterials with internal power sources. We fabricate two active metamaterial structures that, despite still having their power sources implemented externally for reasons of practical convenience, provide evidence in support of the general concept. Effective values are readily achieved at room temperature and pressure and are tunable

throughout the range of stability $-1 < \chi^{(0)} < 0$, resulting in experimentally-determined magnitudes that are over one thousand times greater than those predicted previously. Since $\chi^{(0)} < 0$ is the missing electric analog of diamagnetism, this work opens the door to new technological capabilities such as stable electrostatic levitation.

A key enabler of technological advancement is the development of materials with new or enhanced properties. In this regard, the concept of metamaterials has proven to be fruitful; most famously, it was known since the 1960s that there was no fundamental reason why materials could not exhibit negative refractive index,^[1] yet it was not until the realization of the metamaterials concept around the turn of the century that this property was achieved experimentally.^[2] Hand-in-hand with this breakthrough came new technological advances such as the ‘perfect lens’^[3] and electromagnetic cloaking devices.^[4, 5] In a similar vein, we explore in this paper whether the concept of active metamaterials may be applied to realize another material property that has been discussed theoretically (to a certain extent)^[6, 7] but never developed fully or observed experimentally: ‘true’ negative static electric susceptibility. The primary motivation for pursuing this property is that, as the missing electric analog of diamagnetism, it may be expected to facilitate new technological capabilities. We conclude that true negative static electric susceptibility is possible in active materials, and indicate the practical, materials engineering approaches that can be developed further towards technological application.

Although static magnetic susceptibility may take positive or negative values in paramagnetic and diamagnetic materials respectively, a well-established theoretical argument by Landau et al.^[8] suggests that static electric susceptibility must be positive in “all bodies”. More precisely, the real scalar describing the static electric susceptibility of an isotropic material must be positive, and, in general, the real symmetric second-rank tensor describing the static electric susceptibility must exhibit positive values for all three of its principal components. An equivalent statement is that the presence of a material in the electric field of two conductors always increases, never decreases, their static mutual capacitance when compared to vacuum, regardless of the shape or orientation of the piece of material. There is no reason

to suppose that Landau et al.'s argument does not apply as rigorously to metamaterials as it does to any other type of material. Indeed, using alternative reasoning, Wood and Pendry arrive at the same conclusion when considering metamaterials.^[9] However, it must be borne in mind that, since such arguments implicitly assume that the material is in thermodynamic equilibrium, they do not necessarily hold for materials that are not in thermodynamic equilibrium, as noted, for example, in refs. [6, 7].

It is reasonable to ask whether active materials may violate the restriction, or whether they may nevertheless be subject to the same restriction (perhaps based on more-general theoretical arguments). It is certainly not the case that the static electric susceptibility may take any value whatsoever, if only the material is active. A general and apparently quite rigorous lower bound for the static electric susceptibility is provided by an elementary consideration of an electrical circuit consisting of a dc voltage source, a resistor, and a capacitor in series: a hypothetical material with true negative static dielectric permittivity (static electric susceptibility less than minus one) would, when placed in the capacitor, lead to unphysical instabilities in the quasi-static limit (see, e.g., ref. [10] for a discussion in the context of dispersion in active and passive metamaterials). As a result, static electric susceptibility values less than minus one appear to be ruled out whether the material is in thermodynamic equilibrium or not. The question of negative static electric susceptibility values greater than minus one but less than zero is not addressed by such arguments, and it is negative values in this range that we consider here. The possibility of negative static electric susceptibility in metastable systems with inverted populations has been discussed tentatively by Sanders^[6, 11] and predicted unequivocally by Chiao et al.^[7, 12-14]; the latter authors provide a quantitative estimate for the value of the static electric susceptibility of $\chi^{(0)} = -3.15 \times 10^{-4}$ at a pressure of 1 Torr and a temperature of 180 K in ammonia gas pumped by a carbon

dioxide laser^[12, 13]. However, this scenario may reasonably be considered a negative static electric susceptibility state that is induced in the ammonia gas by the pump laser, as opposed to a material with intrinsic negative static electric susceptibility. Further, and perhaps moreover, the prediction has never been experimentally verified.

We note that the real parts of the principal components of the complex electric susceptibility tensor may readily take negative values for periodic fields, associated with the phase difference between the electric field and the electric polarization, and we emphasize that our interest here is, in contrast, purely in the static case. We also note that we consider the linear electric susceptibility (polarization proportional to electric field) as it pertains to a nonrelativistic, macroscopic, and homogeneous sample of material under the action of an electric field created by external charges. This may be considered the conventional interpretation of the electric susceptibility and is, for example, consistent with the meaning ascribed to the term by Landau et al. and with the definition of the relative permittivity (via $\varepsilon = \chi + 1$) according to current ASTM standards.^[15] There are a number of instances where negative static electric susceptibility or permittivity have been discussed in the literature in relation to quantities that do not correspond to this interpretation; for example, Kirzhnits et al. have shown that static permittivity may be negative in the sense that, if spatial dispersion is taken into account, the longitudinal permittivity at zero frequency but nonzero wavevector may exhibit negative values.^[16] However, Kirzhnits et al.'s scenario concerns the situation where charge sources are placed within the material itself and, for the case of external test electrodes, Kirzhnits et al. reaffirm the conclusions of Landau et al. Herein we seek a true negative static electric susceptibility in a conventional sense, but propose the use of unconventional materials to achieve it, viz. active metamaterials. It has previously been established that active metamaterials may exhibit electromagnetic wave behavior not possible

in their passive counterparts (e.g., ref. [17]), and the concept of metamaterials has been extended to low-frequency or static magnetic fields (e.g., refs. [9, 18]). Our novelty is in applying the concept of active metamaterials to generate a new *electrostatic* material property.

The challenge is to create a material that polarizes in essentially the opposite direction to normal under the action of a static electric field. For simplicity, we focus on creating an anisotropic material for which one principal component, $\chi_z^{(0)}$, of the effective static susceptibility tensor is negative, i.e., for which the induced polarization is in the opposite direction to an electric field applied along the $\pm z$ -axis. The general design concept is as follows: each unit cell, or ‘meta-atom’, consists of: (1) a mechanism to ‘detect’ the local electric field, (2) a system of conductors that may be charged to create an artificial dipole in a direction opposite to that of the electric field, (3) a mechanism by which the conductors may be charged in proportion to the detected electric field, and (4) a means by which to supply the energy necessary to do so. If the magnitude of the artificial dipole due to the charged conductors is sufficiently large with respect to the natural polarization of the materials from which the meta-atom is made, the meta-atom will exhibit a net dipole moment in the opposite direction to normal and have a negative net electric polarizability. On a sufficiently large length-scale, many such meta-atoms may be considered a homogeneous material with $\chi_z^{(0)} < 0$. Given that all materials are inhomogeneous on an atomic length-scale, the description of the macroscopic behavior of a macroscopic sample of such a metamaterial using the quantity $\chi_z^{(0)}$ is no less rigorous than for any other, ‘conventional’, material. Further, it may reasonably be considered a material with the *intrinsic* property $\chi_z^{(0)} < 0$, in the sense that, if it were cut into two pieces (that both still contain many meta-atoms), each piece would still exhibit the same value of $\chi_z^{(0)}$.

Here we report on two experimental implementations that provide evidence in support of this concept (though they fall short of realizing it fully). In the first implementation, a metamaterial structure composed of a 15×15 array of meta-atoms with uniform and externally-controlled artificial polarization was fabricated using etched copper-clad epoxy laminate boards, as shown in **Figure 1a,b** (see also the Supporting Information for a full specification). Each meta-atom contains two copper discs that may be charged by applying an artificial potential difference V_p across them, creating an artificial dipole moment in the $\pm z$ direction. If we choose to apply V_p in proportion to the potential difference V_e across the external test electrodes such that $V_p = \alpha V_e$, we create artificial dipoles in the material whose response mimics natural linear electric polarization, except with a readily-tunable polarizability that is dependent on the value of α .

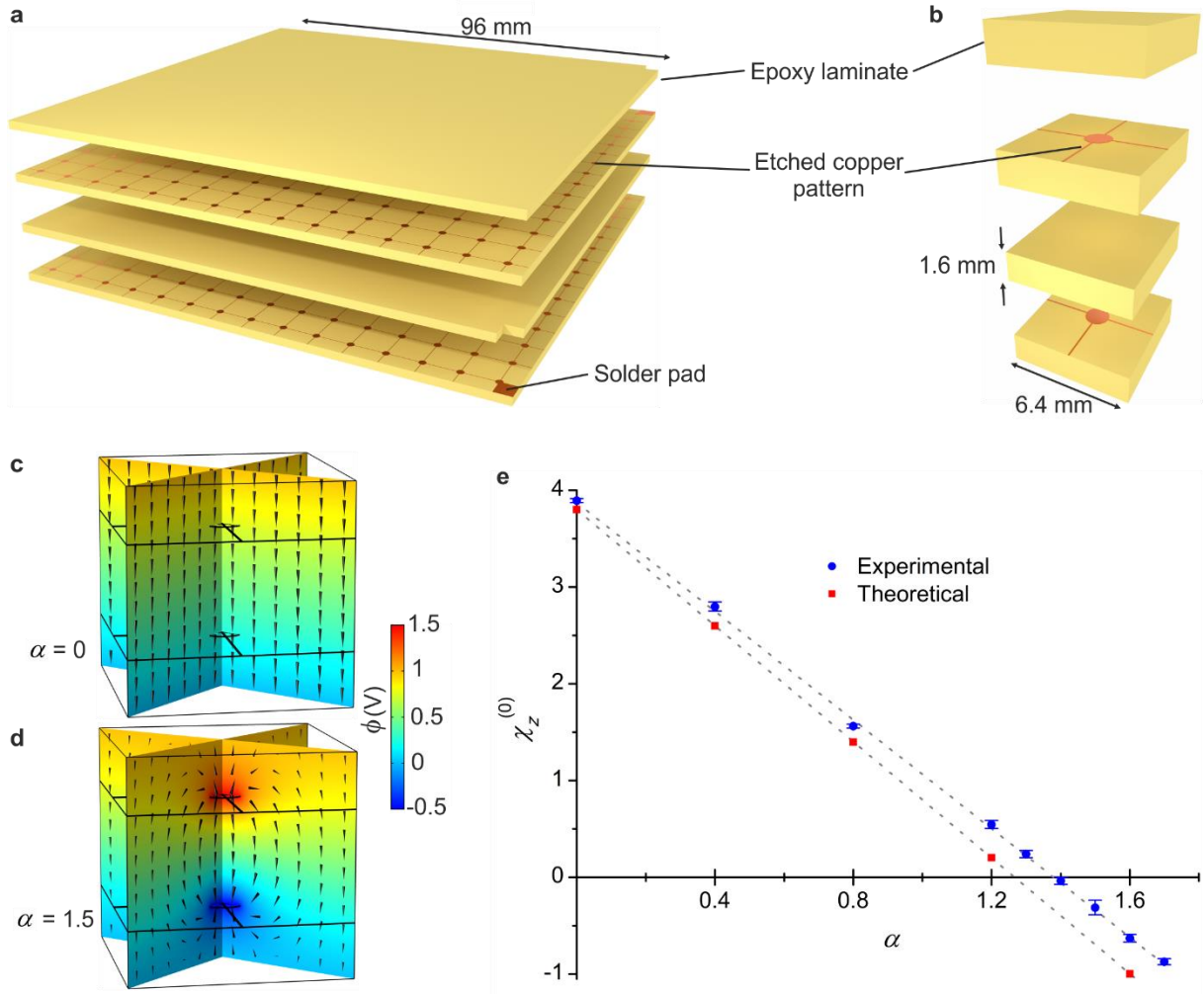


Figure 1. First metamaterial implementation. Exploded view schematic diagrams of: a) the 15×15 metamaterial array formed using stacked epoxy laminate boards with etched copper, and, b) the structure of a single meta-atom. c,d) Theoretical simulations showing the electric potential ϕ and the electric field (black cones) within a meta-atom for reduced driving voltages of $\alpha = 0$ and $\alpha = 1.5$ respectively. The latter exhibits an artificial dipole moment due to the charge on the copper discs and corresponds to a $\chi_z^{(0)} < 0$ state. e) Experimental and theoretical data showing that $\chi_z^{(0)}$ decreases linearly with α and obtains negative values for sufficiently large α . Experimental data points represent the mean of four repeated measurements and error bars represent one standard deviation uncertainties on the mean.

$\chi_z^{(0)}$ was determined via static capacitance measurements on the external electrodes. Care was taken to employ a true dc method rather than simply an ac method at low frequencies (Supporting Information). Results for various values of α are plotted in Figure 1e, from which it is clear that $\chi_z^{(0)}$ decreases linearly with α and can attain negative values for sufficiently large α : that is, for an inverted, artificial, polarization of sufficiently large magnitude.

These experimental results are readily supported by theoretical modelling (Supporting Information). For $\alpha = 0$, i.e., for no artificial dipole applied, the metamaterial behaves essentially as a homogeneous slab of epoxy laminate since the copper discs and tracks are at their ‘natural’ potentials and constitute a negligible volume of the meta-atom. In this case, the modelled electric field within the meta-atom may be seen to be essentially uniform, Figure 1c, and the predicted, positive value of $\chi_z^{(0)}$ is essentially that of the bulk FR4 epoxy laminate itself. For $\alpha > 0$, the artificial dipole is apparent in theoretical plots of the electric potential and electric field, Figure 1d, and the theoretical value of $\chi_z^{(0)}$ is reduced, Figure 1e. The origin of the quantitative discrepancy between the theoretical and experimental data that is apparent in Figure 1e is currently unknown. However, it is clear that the theoretical model supports the qualitative features of the experimental results and, in particular, corroborates the existence of negative static permittivity for sufficiently large α .

While the above implementation provides some evidence in support of the general concept, it cannot be considered a material in any reasonable sense—not least, because the active voltage must be chosen and applied by hand. In a second implementation, we created a meta-atom that responds autonomously to the external electric field, **Figure 2** (see also the Supporting Information for a full specification). It employs the simplest possible field-sensing element: two conductors, the ‘sense electrodes’, which, when subjected to a vertical

electric field produced by the external electrodes, are naturally raised to different electric potentials. The potential difference across the sense electrodes V_s is detected by an instrumentation amplifier of gain G which, in response, applies an amplified potential difference $G \times V_s$ across the drive electrodes. In other words, the sense electrodes and instrumentation amplifier constitute an electric field sensor that acts to feedback and control the potential difference across the drive electrodes.

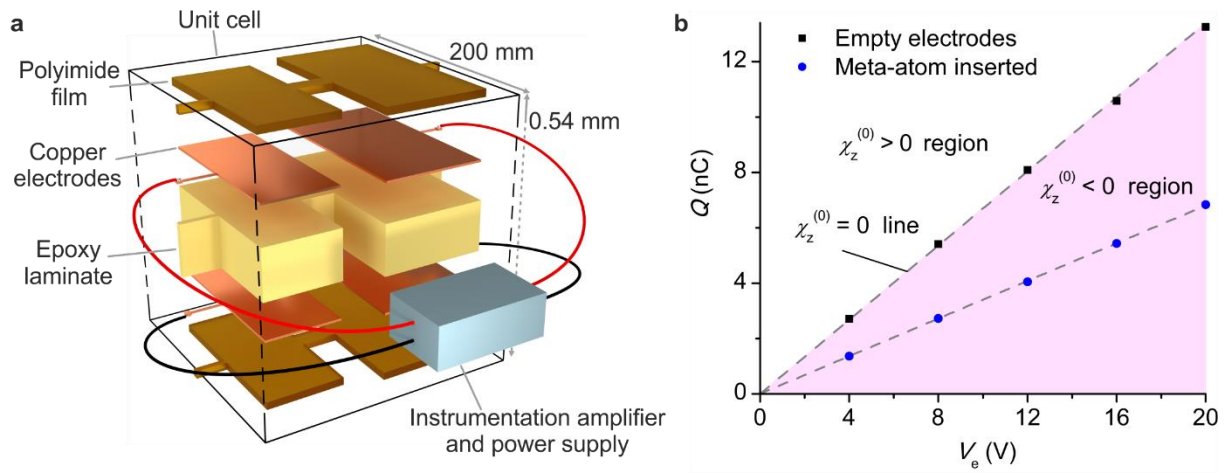


Figure 2. Second metamaterial implementation. a) Exploded view schematic diagram of the meta-atom. The two copper electrodes on the left constitute the sense electrodes and the two on the right the drive electrodes. Relative vertical thicknesses of components within the unit cell are drawn $\times 100$ for clarity. b) Experimental data for the charge Q on the external test electrodes as a function of potential difference V_e applied to them for the empty test electrodes (air as electric), and with the meta-atom inserted. Data points represent the mean of four repeated measurements. One standard deviation uncertainties on the mean values are in the range 0.01–0.04 nC, which is within the thickness of the data symbols.

For this second implementation, $\chi_z^{(0)}$ was again determined experimentally via dc capacitance measurements (Supporting Information). With the meta-atom structure inserted between the external electrodes, the charge Q on the upper external electrode increases linearly with the test voltage V_e , Figure 2b, and the gradient provides a value of $C = (339.9 \pm 0.9)$ pF for the mutual capacitance of the external electrodes. With the structure subsequently removed and the average separation of the external electrodes set to the same value as before, the $Q(V_e)$ data gives the empty capacitance $C_0 = (669.8 \pm 0.6)$ pF, Figure 2b. Thus, it is seen that the mutual capacitance of the test electrodes is reduced by the presence of the structure, and therefore its effective static electric susceptibility must be negative. A value for $\chi_z^{(0)}$ may be calculated most simply via $\chi_z^{(0)} \equiv C/C_0 - 1$, giving the result $\chi_z^{(0)} = -0.49$. The combined statistical and systematic uncertainty on this value is estimated to be 2% (Supporting Information). Here, G was deliberately chosen to produce a value of $\chi_z^{(0)}$ that is approximately in the middle of the region of interest $-1 < \chi_z^{(0)} < 0$. The magnitude of this value is of the order of 10^3 times greater than that predicted theoretically for pumped ammonia gas.^[12,13] By changing G , the value of $\chi_z^{(0)}$ could be readily tuned (Figure S1, Supporting Information).

The second metamaterial implementation again falls short of the general concept in the respect that it consists of only a single meta-atom, and the amplifier and power supply are implemented externally to the unit cell. However, these limitations could be overcome by using integrated chip instrumentation amplifiers and, say, coin cell batteries within the unit cells (though the meta-atom structure would require practical modification and likely require a thicker unit cell to incorporate these components). There should be no problem with the stability of multiple autonomous unit cells for susceptibilities in the range $-1 < \chi_z^{(0)} < 0$. With

appropriate further engineering, it should also be possible to implement materials with more than one negative principal component of the tensor $\chi^{(0)}$, and, in particular, isotropic materials with scalar $\chi^{(0)} < 0$.

We may note, however, that although internal sources of power within each unit cell are required to realize fully the concept of true negative static electric susceptibility, a single external source of power common to each unit cell would likely be preferred in technological applications. Apart from the unique property of reducing the static mutual capacitance of two conductors, demonstrated above, materials with negative static electric susceptibility are expected to exhibit some novel behavior of potential technological interest. For example, negative static electric susceptibility is required for an electromagnetic cloak of the type originally considered by Pendry et al.^[4] that operates for static electric fields. Alternatively, since negative static electric susceptibility materials are, in some sense, the electric analogs of diamagnetic materials, they may be expected to be capable of stable electrostatic levitation in analogy with well-known diamagnetic levitation effects; this may manifest, in principle, either as the levitation of a piece of negative static electric susceptibility material in a static electric field (as considered in refs. [6, 11, 19], and analogous to the levitation of a diamagnetic material in a static magnetic field^[20]), or as the levitation of a ferroelectric body above a piece of negative static electric susceptibility material (as considered in refs. [7, 12] and somewhat analogous to the levitation of a ferromagnet above a superconductor^[21]). Further, a charged body may be expected to be capable of levitation above a piece of negative static electric susceptibility material, with potential application in a new type of particle trap (as proposed in refs. [7, 12]: an effect with no direct magnetic analog due to the absence of magnetic monopoles). Our demonstration of effective negative static electric susceptibility,

with large magnitudes and at room temperature and pressure, brings such possibilities a step closer.

Experimental Section

Experimental details are available in the Supporting Information.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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